Towards a satellite formaldehyde – in situ hybrid estimate for organic aerosol abundance

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Why estimate organic aerosols from satellite OMI HCHO data?

Organic aerosols (OA) : 1. Cloud Condensation Nuclei—Climate

2. Adverse health impact

It is very challenging to model OA

Satellite cannot directly measure OA

In situ: high accuracy; limited spatiotemporal coverage

HCHO & OA: common sources (emissions/secondary productions)

VOCs + OH → …+NO/HO_2 → **HCHO**

VOCs + OH → …+NO/HO_2 → **ROOH/RO_2NO_2/$RONO_2$/R=O/ROH**

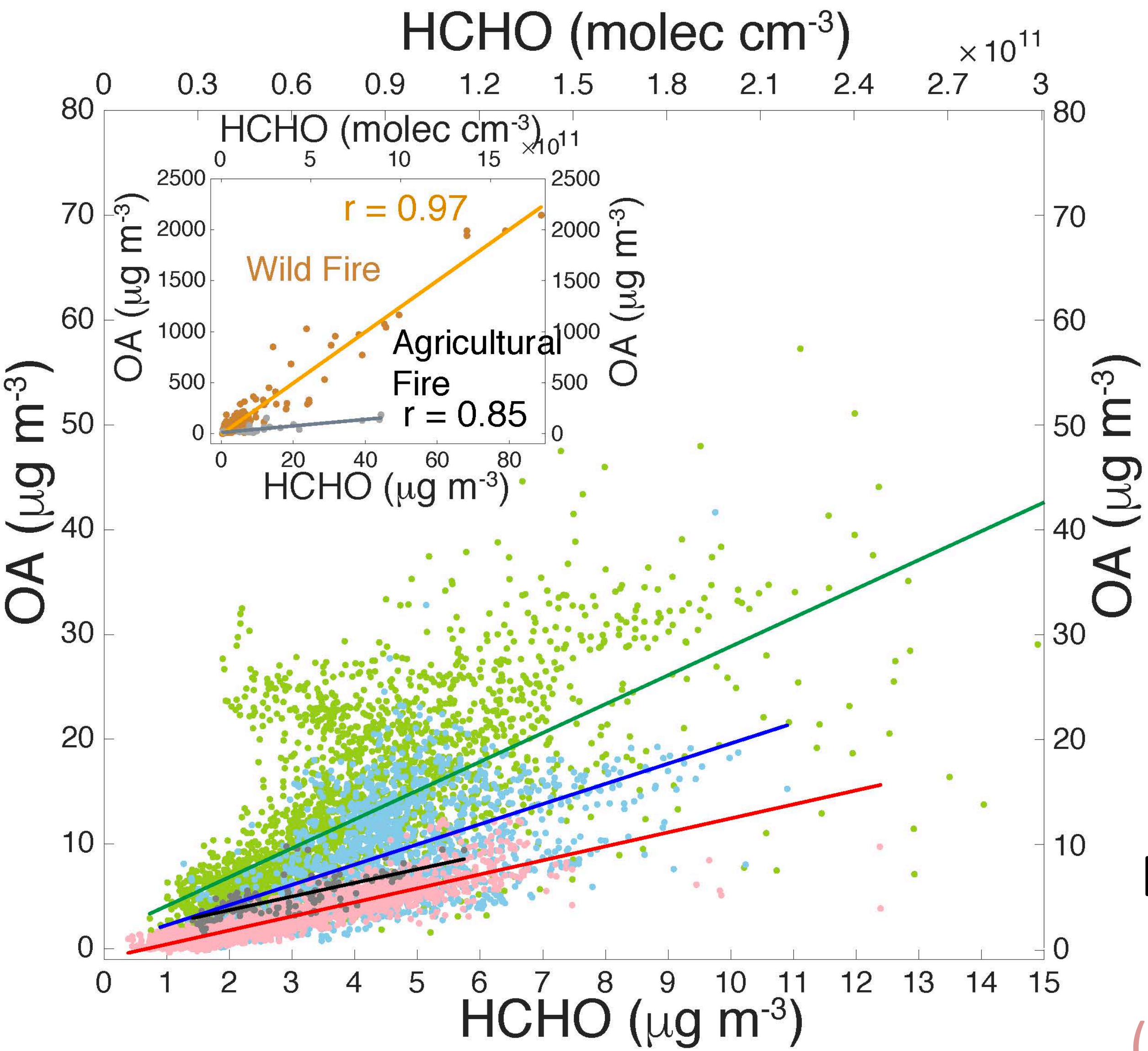
↓ condense

Pre-existing aerosols

secondary organic aerosols

Can we map OA using in situ OA-HCHO relationships and satellite HCHO data?

In situ OA-HCHO depends on the sources



Alt <1 km

Good correlations

KORUS-AQ: r = 0.70
(Heavily polluted Anthropogenic)

SEAC^{4}RS: r = 0.59
(Biogenic)

DC3: r = 0.76 (Biogenic)

CalNex: r = 0.88
(Clean Anthropogenic)

Estimate surface organic aerosols from satellite HCHO data

Datasets

HCHO vertical distributions
e.g., a priori profiles $\eta(i)$

In situ HCHO and OA relationships
$\alpha(i)\ \beta(i)$

Spatiotemporal datasets
e.g., OMI HCHO
$\Omega_{HCHO}(i)$

Ground network measurements
OC → OA

Construct OA estimates

Linear model

$$OA(i) = \Omega_{HCHO}(i)\eta(i)\alpha(i) + \beta(i)$$

Validation

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Mixed-effects model

$$OA = (\alpha+\mu) + (\beta1+\upsilon1)\times\Omega_{HCHO} + (\beta2+\upsilon2)\times\Omega_{NO2} + \beta3\times CO + +\ldots + \varepsilon;$$
$$(\mu,\upsilon1,\upsilon2) \sim \mathcal{N}[(0\ 0\ 0), \Sigma]$$

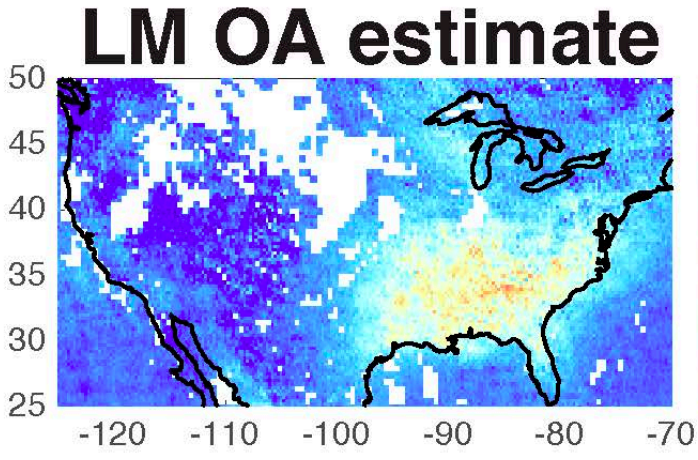
Validation

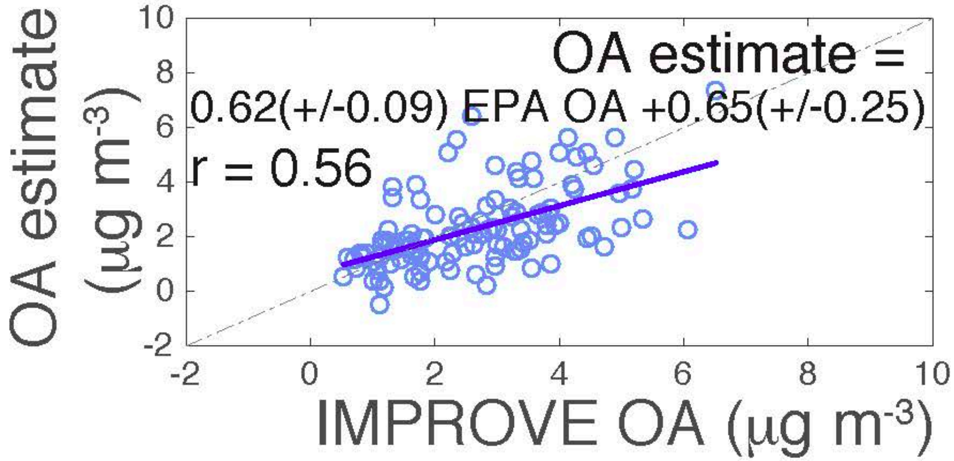
Validation

Cross validation

Simple linear model (LM) OA estimate and IMPROVE sites (rural) comparison

August ,2013





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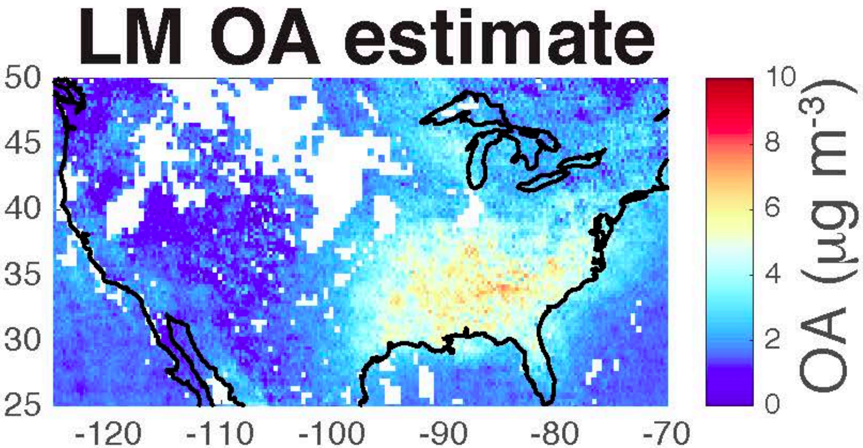
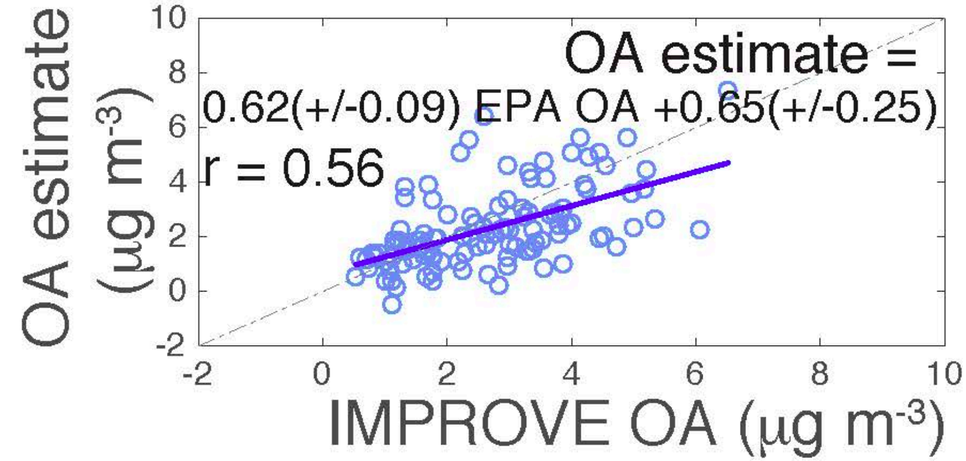
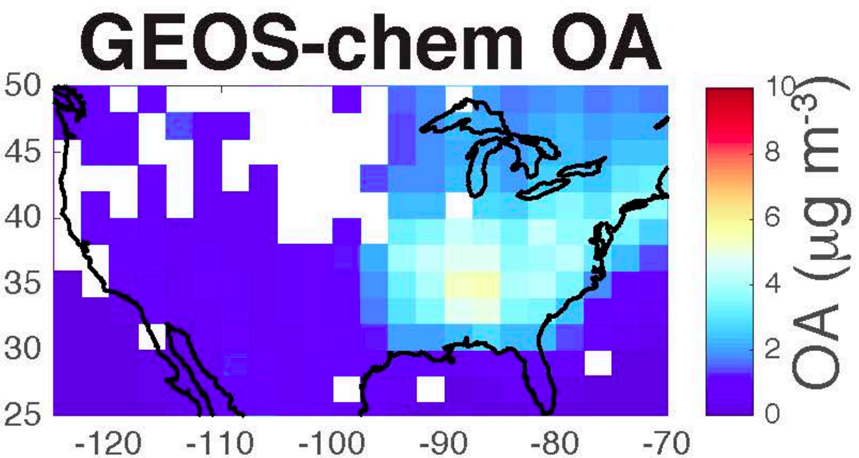
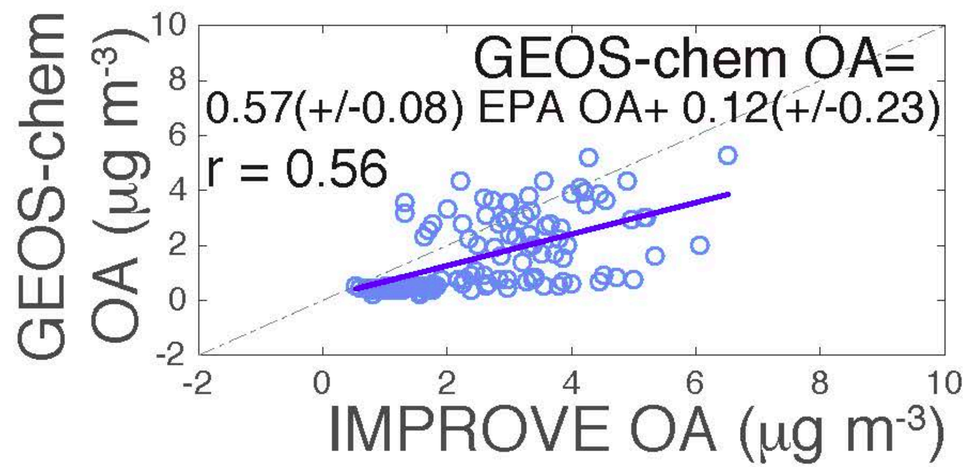






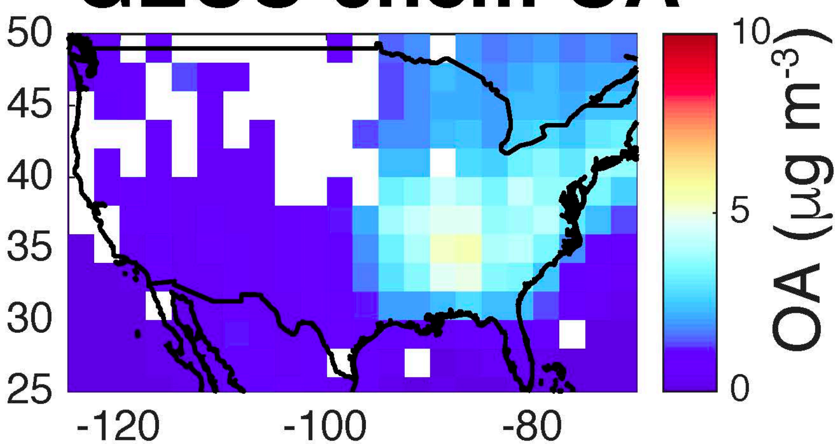


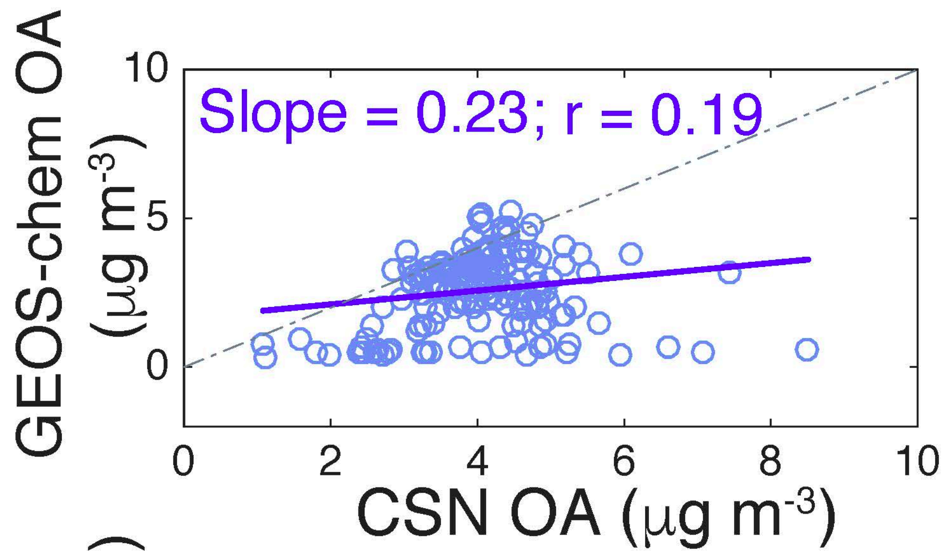
Liao et al., 2019

Mixed-effects model (MEM) OA estimate and CSN sites (urban) comparison

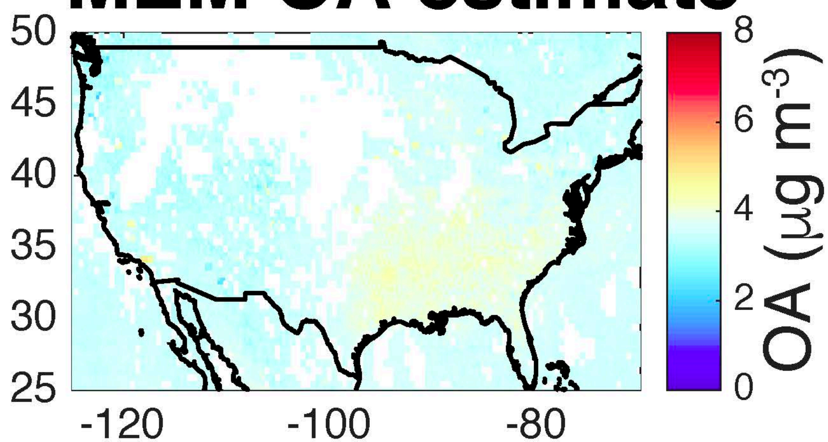
August 2013

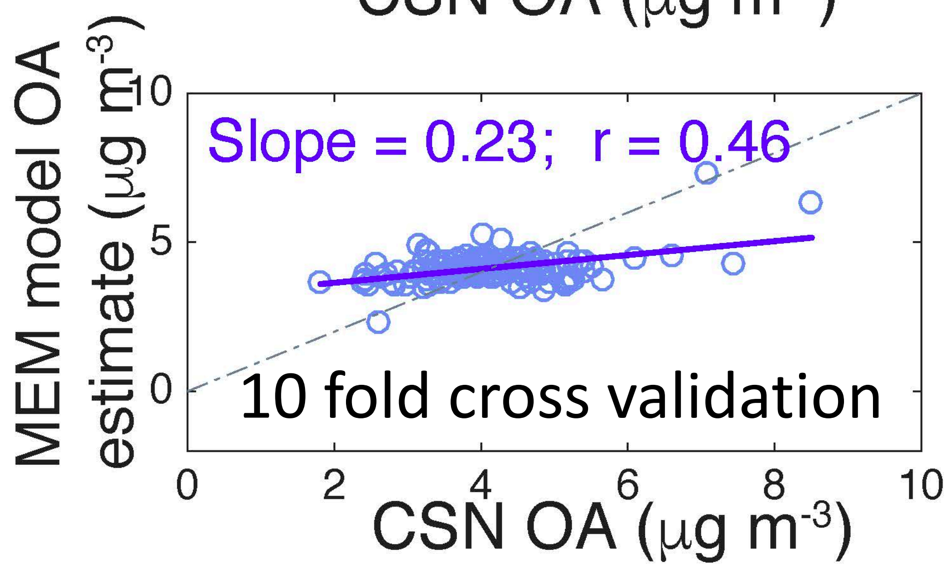
GEOS-chem OA





MEM OA estimate





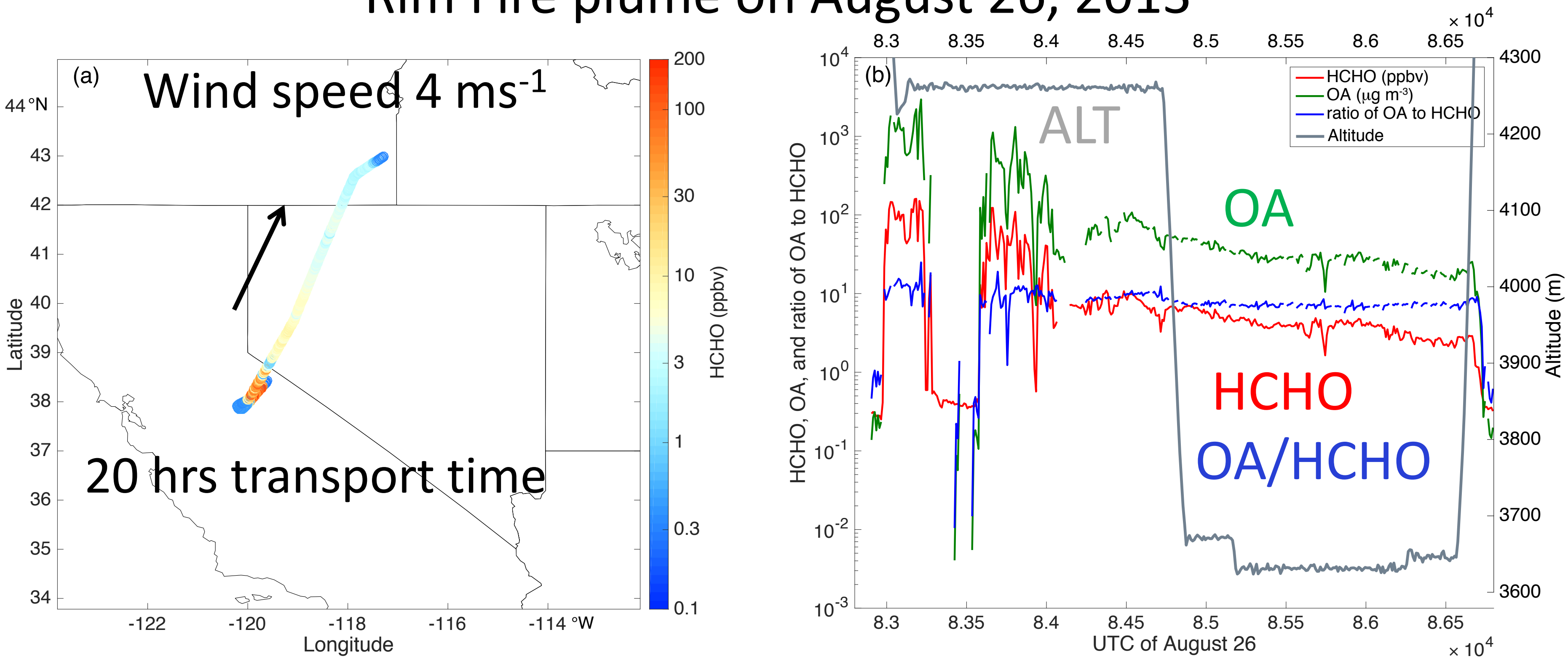
Summary

- Progress toward surface OA mass loadings from satellite HCHO.
- Linear model bulk OA-HCHO relationship: good for rural sites (IMPROVE), better than GEOS-Chem OA. This method can potentially cover regions beyond US where even no ground OA measurements are available.
- Preliminary results: mixed-effects model can obtain an OA estimate map with greater accuracy.

Impact of different lifetimes

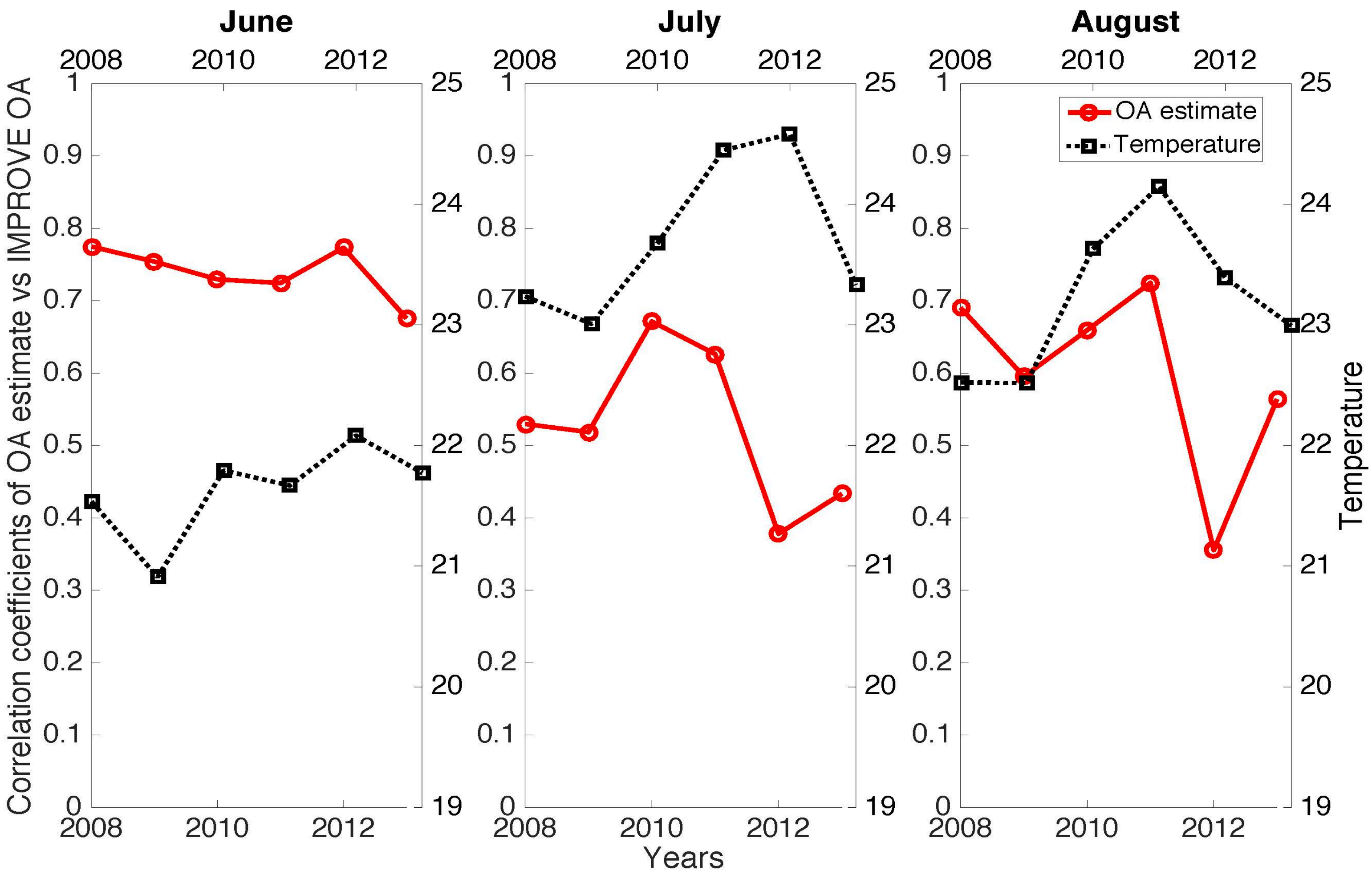
Lifetimes: HCHO (1-3 hrs); OA (~1 week)

Rim Fire plume on August 26, 2013



Nault et al. (2018) has a similar conclusion from KORUS-AQ data. So, in the near field of emissions and chemistry, the ratios of OA/HCHO remain relatively constant.

LM OA estimates for multiple years

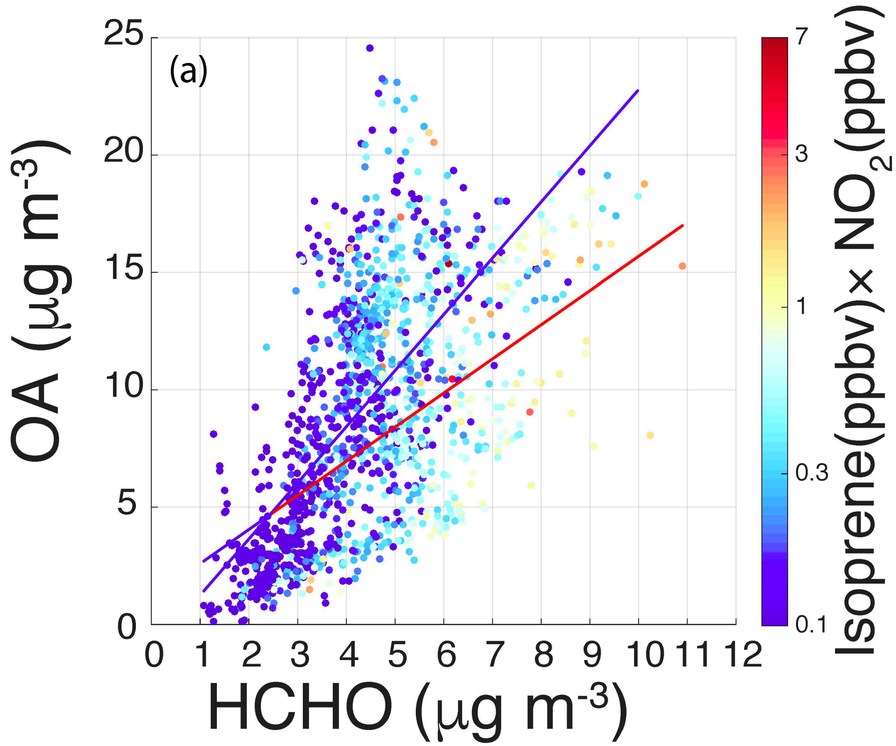


July and August 2012 → severe drought (Seco et al., 2015)?

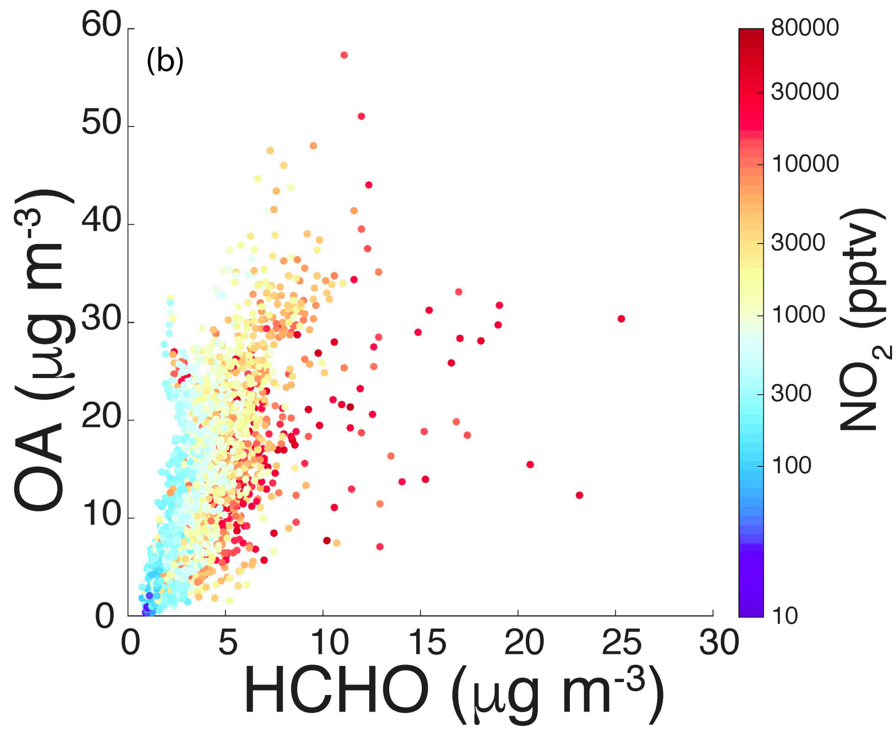
The higher correlation coefficients in June -> related to lower average temperature (less evaporation of OA during filter transport at lower temperature)

OA-HCHO depends on chemical factors

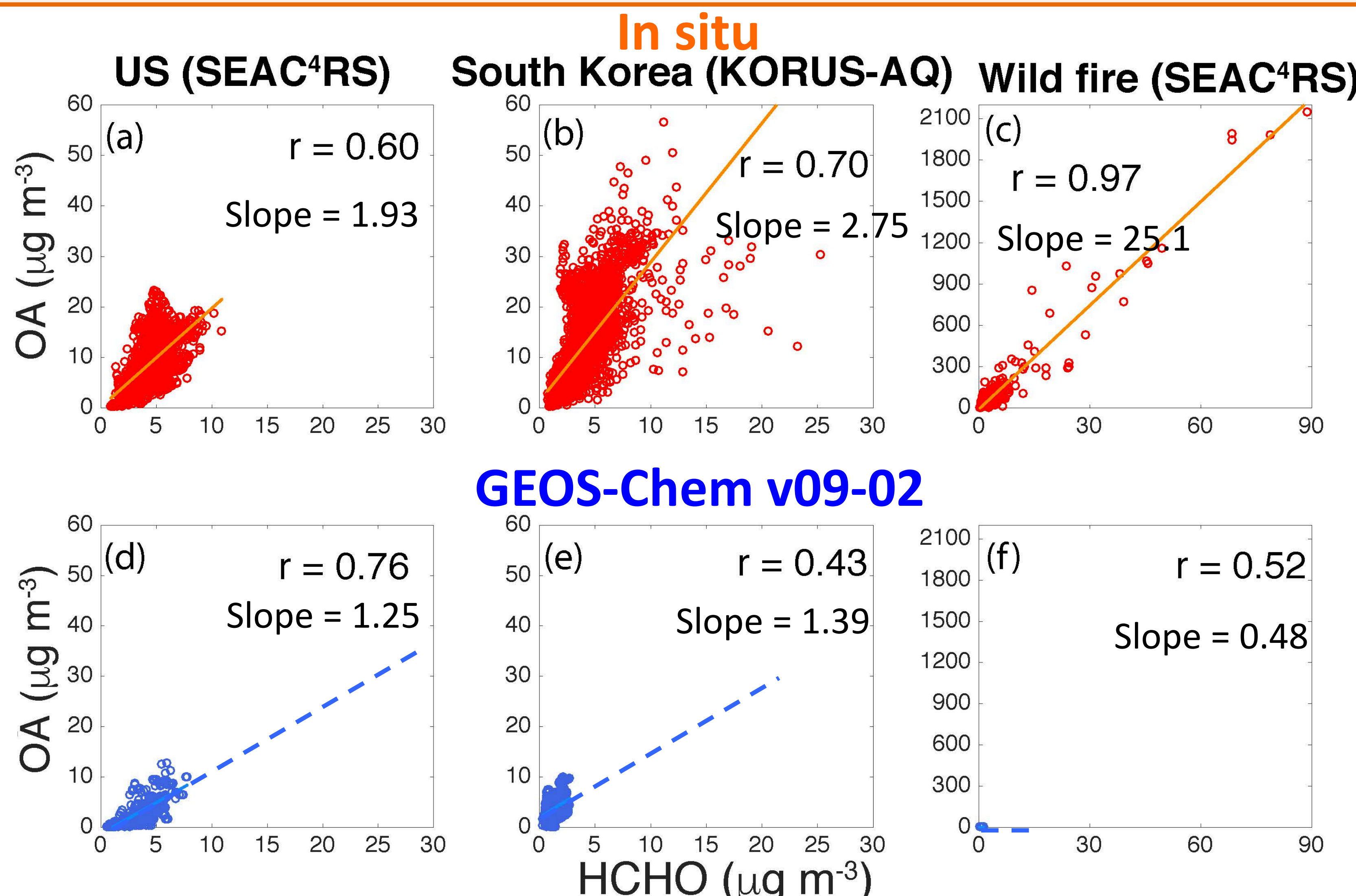
SEAC⁴RS Isoprene and NO_2 Dependence



KORUS-AQ NO_2 Dependence

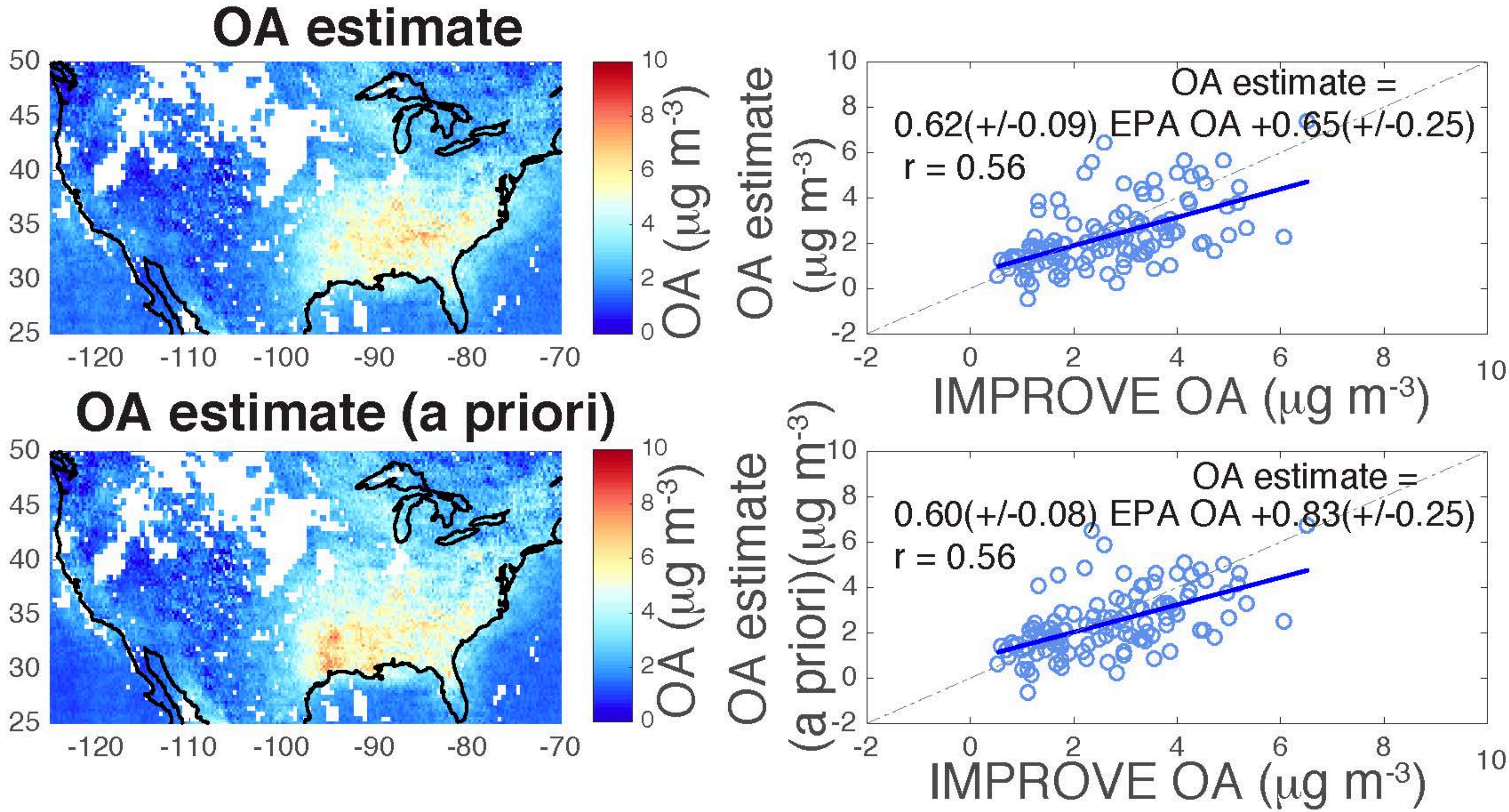


OA-HCHO in situ and modeling



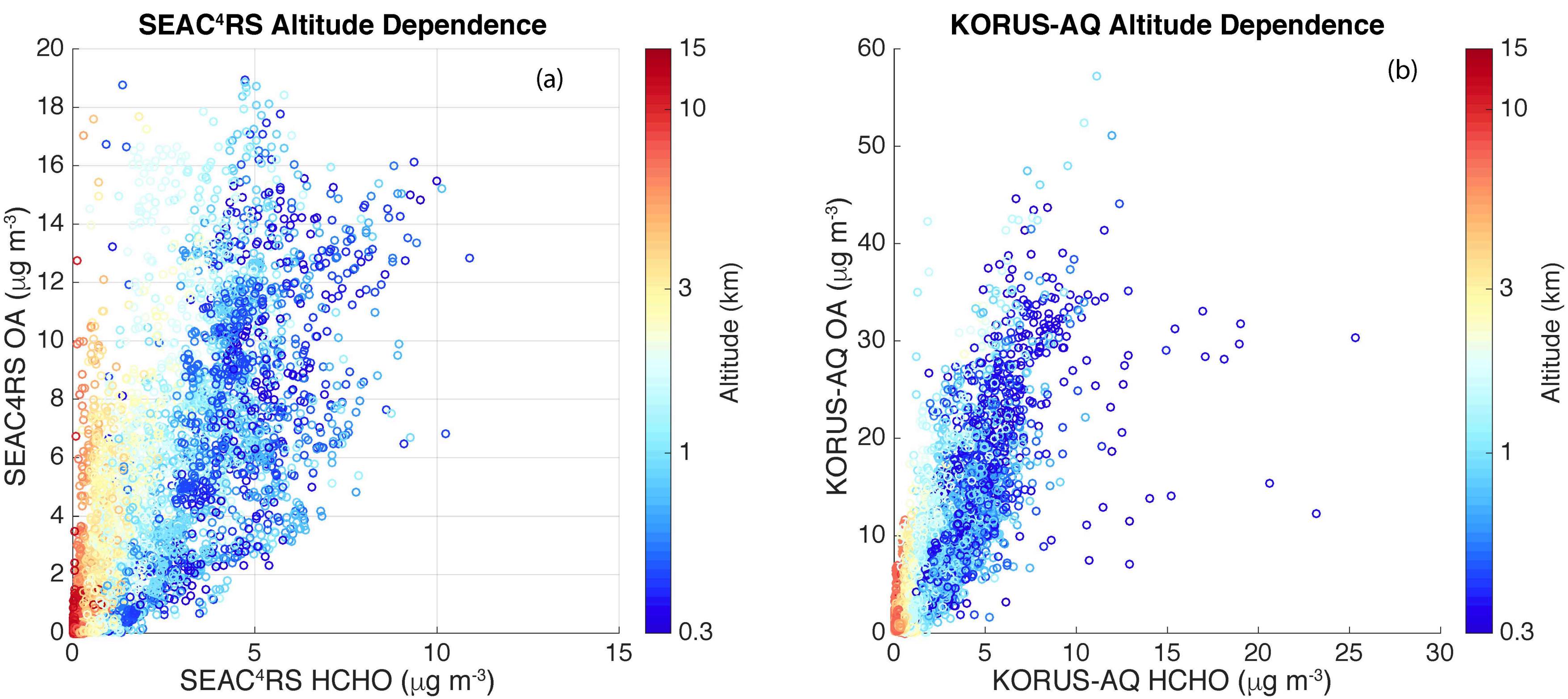
GEOS-Chem still under estimated both OA and HCHO and OA-HCHO ratios, especially for South Korea and biomass burning.

Impacts of different HCHO vertical profiles



Impact of different lifetimes

As expected, OA/HCHO ratio change when air masses are many days aged



Altitude as an indicator of air mass age